

Pseudogap in the Optical Spectra of UPd₂Al₃

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(Received February 6, 2008)

The in-plane optical conductivity of UPd₂Al₃ was measured at temperatures 2 K < T < 300 K in the spectral range from 1 cm⁻¹ to 40 cm⁻¹ (0.14 meV to 5 meV). As the temperature decreases below 25 K a well pronounced pseudogap of 0.2 meV develops in the optical response. In addition we observe a narrow conductivity peak at zero frequency which at 2 K is less than 1 cm⁻¹ wide but which contains only a fraction of the delocalized carriers. The gap in the electronic excitations might be an inherent feature of the heavy fermion ground state.

PACS numbers: 71.27.+a, 74.70.Tx, 78.20.-e

The heavy fermion (HF) compound UPd₂Al₃ is one of the few materials which shows coexistence of superconductivity and magnetic ordering [1]. Susceptibility as well as resistivity data indicate the formation of a coherent state below a characteristic Kondo-lattice temperature $T^* \approx 20$ K; commensurate antiferromagnetic (AF) order develops below $T_N = 14$ K, superconductivity sets in at $T_c = 2$ K. For 2 K < T < 14 K the electronic specific heat shows a $C_{el}/T \propto T^2$ dependence; the effective mass of the charge carriers is estimated as $m^*/m_0 \approx 50$ [1,2] indicating strongly correlated electronic states in coexistence with the long-range AF order. It was suggested [3] that two different groups of uranium 5f states exist in UPd₂Al₃: one group is assumed to be rather localized and responsible for the magnetic properties whereas the states of another sort are less localized and responsible for the superconductivity of the compound.

Optical experiments have proven to be sensitive to the formation of heavy quasiparticles [4]. In the HF compounds above T^* the optical conductivity reveals a broad Drude behavior which characterizes normal metals. Below T^* the increase in the density of states (DOS) at low energies is commonly described by the formation of a narrow Drude-like response with a renormalized scattering rate Γ^* [5]. The signature of magnetic ordering in the optical response of heavy fermions was studied in Ref. [6]. In spin-density-wave (SDW) systems like URu₂Si₂, the opening of a single-particle gap in the electronic DOS at the Fermi surface is clearly seen in the optical properties [6,7]. No effect of the magnetic ordering was found in the infrared response of UPd₂Al₃ so far. Neutron scattering results [8] reveal rather large ordered moments which reside predominantly at the uranium sites. Based on this experimental evidence, UPd₂Al₃ was described as a local-moment magnet and hence, the magnetic ordering should have only a minor influence on the electronic DOS. In this Letter we report on optical experiments at millimeter-submillimeter (mm-submm) wavelengths which clearly show the development of a pseudogap in the DOS in UPd₂Al₃ below 25 K; our preliminary

study on another film [9] showed the same features in the optical response; these findings cannot be easily explained by existing models.

The highly c -axis oriented thin (150 nm) film of UPd₂Al₃ was prepared on (111) oriented LaAlO₃ substrate (thickness 0.924 mm) by electron-beam co-evaporation of the constituent elements in a molecular-beam epitaxy system [10]. The phase purity and structure of the film were investigated by X-ray and reflection high-energy electron diffraction. The high quality of the film is seen in dc resistivity data displayed in Fig. 1 which perfectly agree with measurements of single crystals [1].

For the measurements a coherent source mm-submm spectrometer was employed [11] utilizing a set of backward wave oscillators as monochromatic but continuously tunable sources. The use of a Mach-Zehnder arrangement allows for measuring both the amplitude and the phase of the signal transmitted through the sample, which in our case is a film on a substrate (with the electric field \mathbf{E} perpendicular to the c -axis). From these two quantities the conductivity $\sigma(\omega)$ and dielectric constant $\epsilon(\omega)$ of the film are evaluated using Fresnel's formulas for a two-layer system [12] without assuming any particular model. The optical parameters of the substrate are determined beforehand by performing the experiments on a blank substrate. The large size of the sample (1×1 cm²) allowed us to extend the measurements to very low frequencies, from $\omega/(2\pi c) = 40$ cm⁻¹ down to 1.15 cm⁻¹. In addition, we performed microwave experiments at 10 GHz using a setup designed for measuring superconducting films [13]. A small slice of a sample (4×1 mm²) was placed in the electric field maximum of a cylindrical cavity ($\mathbf{E} \perp c$); we obtain the conductivity (Fig. 1) and dielectric constant from the change in width and shift in frequency using cavity perturbation theory.

In Fig. 2 we present the raw transmission spectra for the UPd₂Al₃ film on the substrate for two selected temperatures (100 K and 2 K) which illustrate clearly our main finding. The fringes in the spectra are due to multi-reflection of the radiation within the plane-

parallel substrate acting as a Fabry-Perot resonator for the monochromatic radiation [12]. The distance between the peaks is mainly determined by the thickness and refractive index of the substrate, their amplitude by the parameters of the film. At $T \geq 30$ K the transmission maxima and minima only slightly depend upon frequency, implying a frequency independent conductivity and dielectric constant of the UPd₂Al₃ film. In contrast, the overall transmission for lower temperatures is strongly reduced below 10 cm^{-1} due to absorption within the film. At lower frequencies, below 3.5 cm^{-1} , the transmitted signal increases again, indicating an absorption edge. This feature can also be seen by looking at the temperature dependencies of the transmission and the conductivity as plotted in Fig. 1 for several fixed frequencies.

The conductivity and dielectric constant of UPd₂Al₃ are plotted in Fig. 3 as functions of frequency for different temperatures. Within our accuracy they are frequency independent for $T \geq 30$ K which resembles a low-frequency response of a Drude metal; the value of the conductivity agrees well with the dc result. It is clearly seen that for low temperatures the optical response of UPd₂Al₃ differs from that of a renormalized Drude metal. The simple Hagen-Rubens relation in general used to extrapolate the far-infrared reflectivity below 30 cm^{-1} [4] totally misses the two features in the mm-submm range. First, the optical conductivity $\sigma(\omega)$ clearly shows the development of a minimum below 3 cm^{-1} at $T < 30$ K with a correspondent pronounced increase of $\epsilon(\omega)$. We ascribe this minimum to the opening of a pseudogap in the electronic DOS (see below). This feature gradually disappears with increasing temperature and is not seen above 30 K. Second, at frequencies below approximately 1.5 cm^{-1} the conductivity increases towards considerably higher dc values leading to a very narrow peak at $\omega = 0$. With the help of our microwave data, we estimate its width to be 0.3 cm^{-1} at $T = 2$ K.

To discuss our findings let us turn back to Fig. 1. Below T_N the temperature dependence of the dc resistivity of the film can be described using the expression for an antiferromagnet with energy gap E_g [2,14]:

$$\rho(T) = \rho_0 + aT^2 + bT \left(1 + \frac{2k_B T}{E_g} \right) \exp \left\{ -\frac{E_g}{k_B T} \right\}. \quad (1)$$

Here ρ_0 gives the residual resistivity and the second term indicates the electron-electron scattering. Fitting the dc data of Fig. 2 yields a gap value $E_g = 1.9 \text{ meV}$, which corresponds to the one reported in [2,10,14]. From point contact measurements a larger gap up to 12.4 meV was suggested [15]. Using thin films of UPd₂Al₃ recent tunneling experiments in the superconducting and normal state [16] clearly demonstrate that a reduction of the DOS remains in the temperature range roughly up to T_N . The existence of a gap in either the electronic DOS or in the magnon spectrum was opposed by Caspary *et*

al. [3] and on the ground of infrared measurements by Degiorgi *et al.* [6]. In the light of the low-energy measurements presented in this Letter the arguments will be reconsidered below.

To analyze the spectral weight of the optical response, we first neglect the pseudogap and describe the mm-submm spectra of $\sigma(\omega)$ and $\epsilon(\omega)$ in terms of a renormalized Drude response (dotted line in Fig. 4)

$$\sigma(\omega) = \frac{\Gamma^*}{4\pi} \frac{(\omega_p^*)^2}{\omega^2 + (\Gamma^*)^2}, \quad \epsilon(\omega) = 1 - \frac{(\omega_p^*)^2}{\omega^2 + (\Gamma^*)^2} \quad (2)$$

where the plasma frequency is related to the carrier density n and the effective mass m^* by $\omega_p^* = \sqrt{4\pi n e^2 / m^*}$. At $T = 2$ K we find $\omega_p^* / (2\pi c) \approx 4300 \text{ cm}^{-1}$. Assuming that the total number of charge carriers remains unchanged, sum-rule arguments give $\omega_p / \omega_p^* = \sqrt{m^* / m_b}$ where ω_p is the unrenormalized plasma frequency and m_b the band mass. With $\omega_p / (2\pi c) = 44000 \text{ cm}^{-1}$ [6] we obtain $m^* / m_b \approx 105$. Using experimental [17] and calculated [18] de Haas-van Alphen spectra we know that approximately 35 % of the electrons contribute to the HF state, leading to the effective mass of $m^* / m_b = 40$.

In contrast to the above description by a renormalized Drude metal, we find clear evidence for a narrow pseudogap which develops in UPd₂Al₃ at low temperatures reducing the spectral weight in the conductivity spectrum and correspondingly the plasma frequency ω_p^* . At 2 K this reduction amounts in 40% (hatched area in Fig.4) leading finally to the effective mass $m^* / m_b = 60$ which agrees well with the value obtained by thermodynamic methods ($m^* / m_0 = 41 - 66$) [1,2], but is a somewhat smaller than those got from previous infrared measurements ($m^* / m_b = 85$) [6]. The gap value can be determined using a semiconductor approach with $\sigma(\omega) \propto \omega^{-1} \sqrt{\hbar\omega - E_g}$ giving $E_g \approx 0.23 \text{ meV}$ (solid line in Fig. 4). This value is essentially temperature independent (Fig. 3a) in contrast to the BCS-like temperature behavior of the energy gap in superconducting or density-wave ground systems. Instead it becomes more and more pronounced as the temperature is lowered.

The most interesting question is the relation between the AF ordering and the pseudogap. The fact that we also see the gap-like feature above T_N up to 30 K does not rule out its connection to the magnetic ordering since an incommensurate phase was observed in UPd₂Al₃ up to $T \approx 20$ K by neutron diffraction experiments [8], and a maximum of the magnetic susceptibility appears around 35 K [1]. We want to discuss three possible explanations: (i) the pseudogap in the optical response may be related to spin-wave excitations; (ii) the formation of a SDW ground state may lead to the opening of an energy gap. (iii) On the other hand since T^* is also in this temperature range, the gap in the electronic DOS might be a property of the coherent HF ground state.

(i) Inelastic neutron scattering experiments found two very-low-energy modes, one at 1.5 meV which is ascribed

to spin-wave excitations, and another at 0.4 meV which is associated with superconductivity [19]. These modes exhibit a strong T and \mathbf{q} -dependence. Besides the resistivity $\rho(T)$ discussed above, the drop of $C_{el}(T)$ [1,3] and torque measurement results of the magnetization [20] were also interpreted as being due to the opening of a gap in the magnetic excitation spectrum. However, we exclude the possibility that the features discovered in our low-temperature mm-submm spectra are due to purely magnetic absorption since we were not able to obtain a reasonable fit of these features by a “magnetic Lorentzian” absorption line. We definitely see the development of a gap in the *electronic* excitation spectra.

(ii) The low-temperature decrease of the resistivity of UPd₂Al₃ can be well described by an exponential behavior [Eq. (1)]. The so-obtained gap $E_g=1.9$ meV may indicate the formation of a SDW state like in URu₂Si₂ [2,14]. However, the gap energy 0.23 meV obtained from our optical results is about an order of magnitude smaller; it is also a factor of 20 below the value one would expect from mean field theory $E_g = 3.5k_B T_N \approx 4.2$ meV. Also the comparison of NMR and NQR results for UPd₂Al₃ [21] and for the SDW systems URu₂Si₂ [22] and UNi₂Al₂ [23] does not corroborate the SDW picture for UPd₂Al₃. The itinerant antiferromagnetism of a SDW also is at odds with the formation of local moment magnetism deduced from susceptibility [1] and neutron scattering [8].

(iii) It was suggested [3,18] that two electronic subsystems coexist in UPd₂Al₃. One of them is a rather localized uranium $5f$ state responsible for the magnetic properties, the other part is delocalized and determines the HF and superconducting properties. From the London penetration depth $\lambda_L(0) = 450$ nm [3] we calculate [24] the plasma frequency of superconducting carriers $(2\pi\lambda_L)^{-1} = 3540$ cm⁻¹ and find a perfect agreement with $\omega_p^*/(2\pi c) = 3500$ cm⁻¹ obtained from the spectral weight under our mm-submm conductivity spectrum just above T_c (grey area in Fig. 4). This not only is an independent confirmation of the pseudogap but it also implies that all carriers seen in our spectra are in the HF ground state and eventually undergo the superconducting transition below T_c . We can definitely rule out an assignment of the gap to the localized carriers of the AF ordered states with the delocalized carriers contributing only to the narrow feature at $\omega = 0$ because with a plasma frequency of 1500 cm⁻¹ at low temperatures, it accounts for only 18% of the carriers which become superconducting. This means that also the excitations above the gap stem from the delocalized states and that the pseudogap observed in our conductivity spectra is either inherent to the heavy quasiparticle state or it is related to magnetic correlations of the second subsystem.

In conclusion, the electrodynamic response of UPd₂Al₃ in the low energy range from 0.14 meV to 5 meV (1 cm⁻¹ to 40 cm⁻¹) exhibits a behavior at low temperatures ($T \leq 25$ K) which cannot be explained within the simple

picture of a renormalized Fermi liquid. We observe an extremely narrow (less than 0.1 meV) Drude-like peak at $\omega = 0$ (dashed line in Fig. 4) and a pseudogap of about 0.2 meV. The experiments yield indications that this pseudogap is not a simple SDW gap but rather is connected to correlations of the delocalized carriers, and may be a general signature of HF compounds. Recent millimeter wave experiments on UPt₃ [25] which show a peak in the conductivity at 6 cm⁻¹ for temperatures below 5 K indicate a similar scenario. While the finite-energy excitations occur at comparable frequency in UPd₂Al₃ and in UPt₃, the energy scale of the magnetic ordering is somewhat different, since $T_N \approx 14$ K in UPd₂Al₃ while magnetic correlations are found in UPt₃ only below 5 K [26]. Investigations of the magnetic field dependence which might shine light on this problem are in progress.

We thank G. Grüner, A. Loidl, M. Mehring and A. Mukhin for helpful discussions. The work was partially supported by the Deutsche Forschungsgemeinschaft (DFG) via Dr228/9 and SFB 252.

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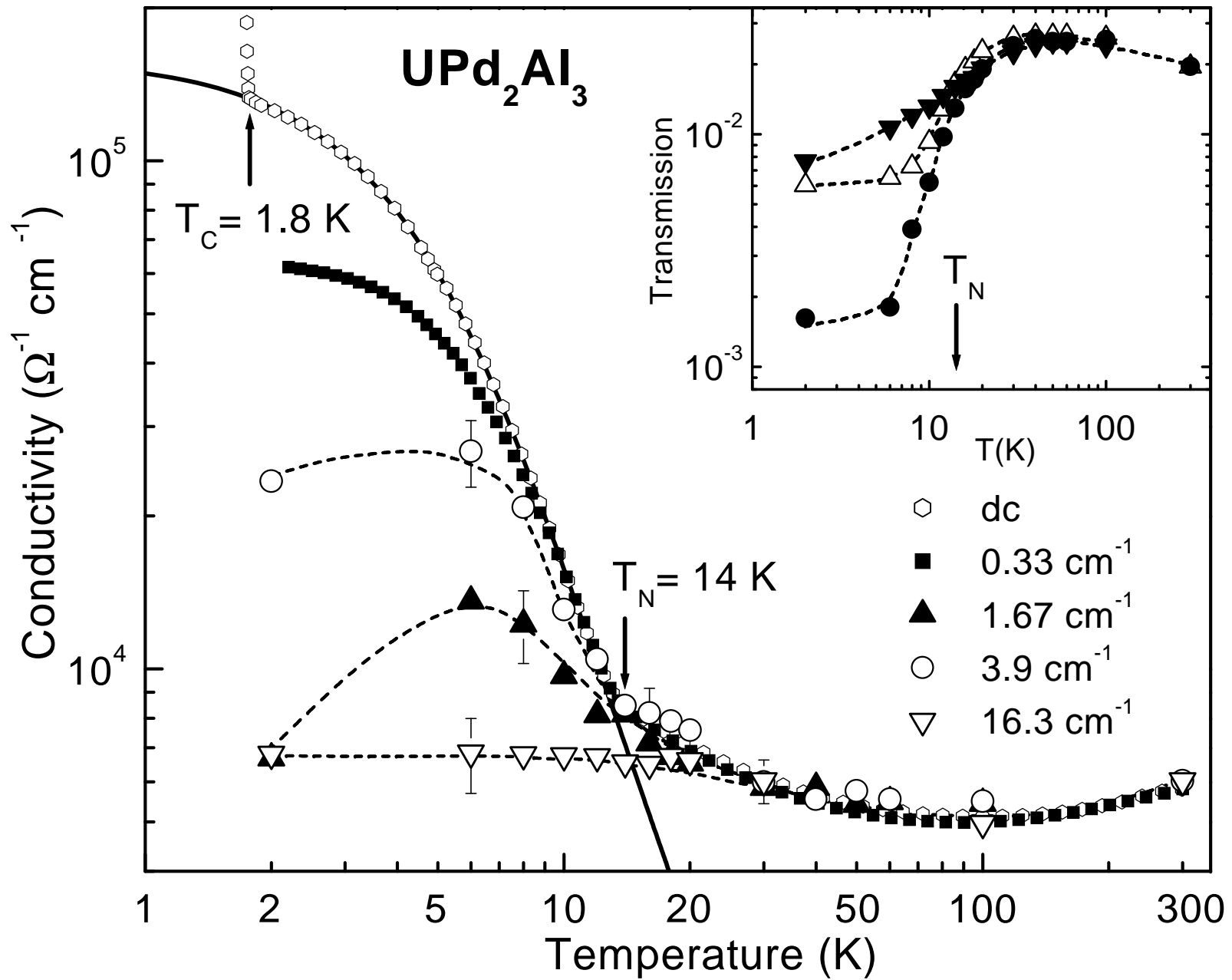
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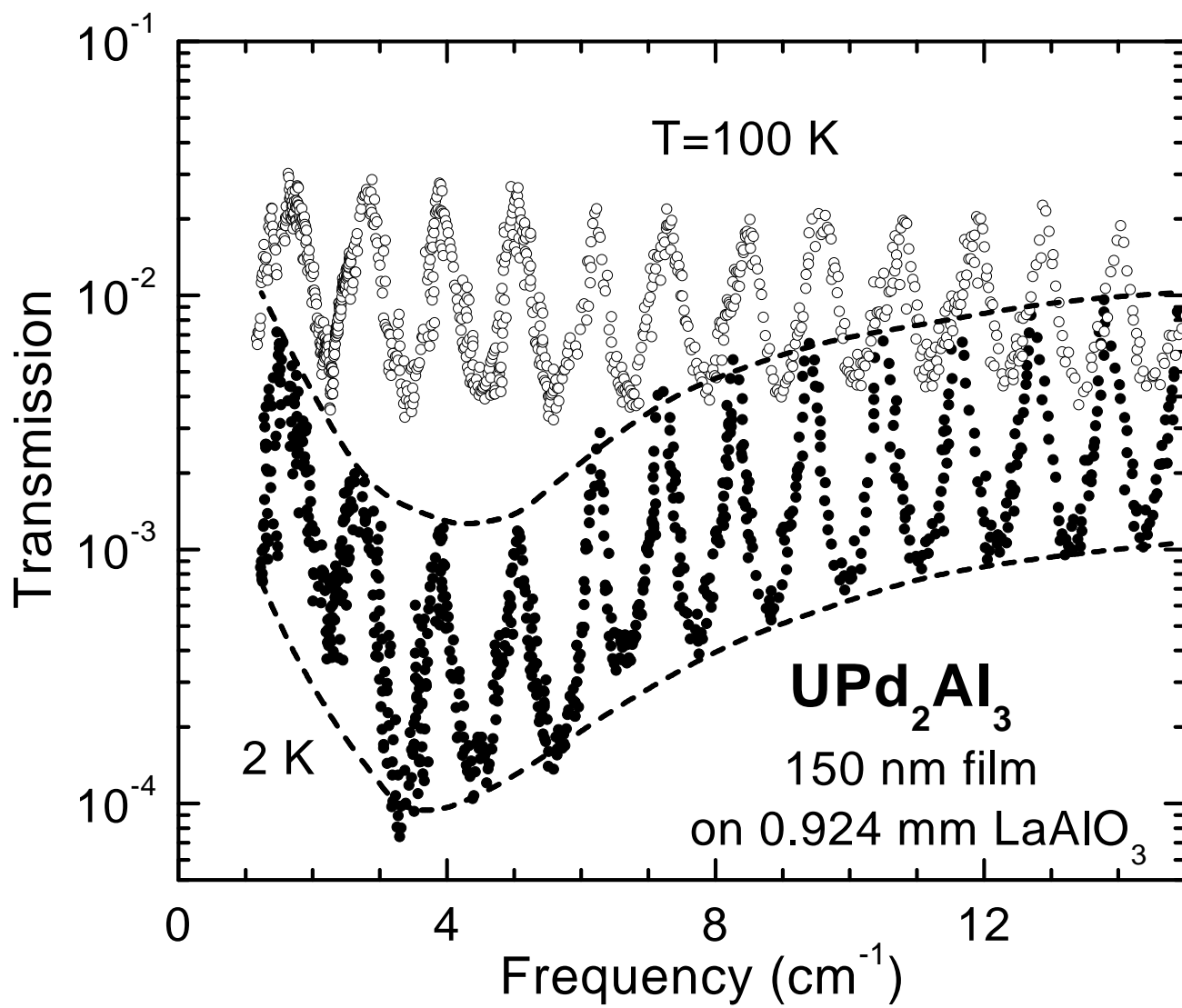
FIG. 1. Temperature dependence of the dc and ac conductivities of a UPd_2Al_3 film. At T_N the system orders antiferromagnetically, at T_c it becomes superconducting. The solid line represents a fit using Eq. (1) with the gap $E_g = 1.9$ meV. The inset shows the temperature dependence of the transmission through film on LaAlO_3 substrate for different frequencies. Dashed lines are guides to the eye.

FIG. 2. Spectra of transmission of a 150 nm thick UPd_2Al_3 film on LaAlO_3 (thickness 0.924 mm) for two temperatures $T = 100$ K and 2 K. The periodical maxima are caused by interference of the radiation in the substrate. The dashed lines connecting minima and maxima are drawn to emphasize the overall frequency dependence of the transmission of the film.

FIG. 3. (a) The conductivity and (b) the dielectric constant of UPd_2Al_3 as functions of frequency at several temperatures. The dc conductivities are shown on the left side of the upper panel; the data at 0.33 cm^{-1} are obtained by microwave cavity measurements. The inset shows the optical conductivity over a wide frequency range using the data of [6]. The lines are drawn to guide the eye.

FIG. 4. Frequency dependent conductivity $\sigma(\omega)$ (dots) of UPd_2Al_3 at $T = 2$ K. The dotted line shows a fit of the data with the renormalized Drude conductivity [Eq. (2)]. The dashed curve is a Drude description of the $\omega = 0$ peak with a width of 0.3 cm^{-1} . The solid line indicates a $\omega^{-1}\sqrt{\hbar\omega - E_g}$ behavior of the pseudogap used for the absorption edge in semiconductors.





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